PCT WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 5 : B32B 27/06, 27/36 — A1

(11) International Publication Number: WO 94/13476

(43) International Publication Date: 23 June 1994 (23.06.94)

(21) International Application Number: PCT/US92/10699

(22) International Filing Date: 9 December 1992 (09.12.92)

(71) Applicants *(for all designated States except US)*: HOECHST AKTIENGESELLSCHAFT [DE/DE]; Postfach 800 320, D-6320 Frankfurt am Main 80 (DE). HOECHST CELANESE CORPORATION [US/US]; Route 202-206 North, Somerville, NJ 08876 (US).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: BENNETT, Cynthia [US/DE]; Mainstrasse 22, D-6200 Wiesbaden (DE). CHOE, E-Won [US/US]; 130 Radtke Road, Randolph, NJ 07869 (US). FLINT, John, Anthony [GB/US]; 150 Lenape Lane, Berkeley Heights, NJ 07922 (US). KUHMANN, Bodo [DE/DE]; Lindenstrasse 5, D-6258 Runkel 5 (DE).

(74) Agents: CLEMENTS, Gregory, N. et al.; Hoechst Celanese Corporation, 4000 Barclay Downs Drive, Patent Department, Charlotte, NC 28232-2414 (US).

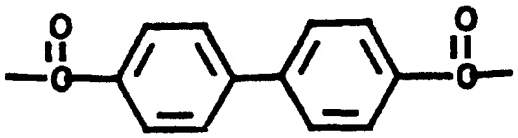
(81) Designated States: CA, JP, KR, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published
With international search report.

(54) Title: COPOLYESTER FILM PRIMED WITH ACRYLIC POLYMERS

(57) Abstract

The invention deals with a primer coated, biaxially oriented self-supporting, mono- or multilayer copolyester film, wherein the copolyester is PENBB and wherein the primer coating contains an acrylic polymer. PENBB is a copolyester containing units of formula (I).



(I)

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	GB	United Kingdom	MR	Mauritania
AU	Australia	GE	Georgia	MW	Malawi
BB	Barbados	GN	Guinea	NE	Niger
BE	Belgium	GR	Greece	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IE	Ireland	NZ	New Zealand
BJ	Benin	IT	Italy	PL	Poland
BR	Brazil	JP	Japan	PT	Portugal
BY	Belarus	KE	Kenya	RO	Romania
CA	Canada	KG	Kyrgyzstan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo			SE	Sweden
CH	Switzerland	KR	Republic of Korea	SI	Slovenia
CI	Côte d'Ivoire	KZ	Kazakhstan	SK	Slovakia
CM	Cameroon	LI	Liechtenstein	SN	Senegal
CN	China	LK	Sri Lanka	TD	Chad
CS	Czechoslovakia	LU	Luxembourg	TG	Togo
CZ	Czech Republic	LV	Latvia	TJ	Tajikistan
DE	Germany	MC	Monaco	TT	Trinidad and Tobago
DK	Denmark	MD	Republic of Moldova	UA	Ukraine
ES	Spain	MG	Madagascar	US	United States of America
FI	Finland	ML	Mali	UZ	Uzbekistan
FR	France	MN	Mongolia	VN	Viet Nam
GA	Gabon				

COPOLYESTER FILM PRIMED WITH ACRYLIC POLYMERS

The present invention relates to biaxially oriented PENBB film coated on one or both sides with acrylic polymers which render the copolyester film surface receptive to additional reprographic or matte coatings applied thereto, and to film containing the reclamation product of such coated film.

Background Of The Invention

Oriented polyester films, particularly biaxially oriented film composed of polyethylene terephthalate (PET), are widely used as a base for drafting film, photographic film, and reprographic film, as well as for packaging and labeling applications.

Polyester films, especially PET films, have disadvantages which still need to be overcome, e.g., UV resistance, dimensional stability, heat stability, and hydrolysis resistance are still poor. Moreover, PET film is hydrophobic and is not readily receptive to coating in most applications where the film is to serve as a base or support for other coatings, and must therefore be first coated on one or both sides with a primer coating which adheres to the film and is receptive as well to other coatings applied to it. For example, U.S. Patent Nos. 2,627,088 and 2,698,240 teach a primer coating for PET film comprising a terpolymer composition of vinylidene chloride, acrylic ester and itaconic acid. This primer layer is said to have excellent adhesion to the polyester surface and to water or alcohol based photographic gelatin layers subsequently coated thereon.

Another known PET film primer includes copolymers of a vinyl halogenoester, such as vinyl chloroacetate which may be copolymerized with numerous different monomers such as acrylic and methacrylic acids, esters and amides, olefins and vinyl alcohols as disclosed in U.S. Patent No. 3,674,531. Such copolymers may also be crosslinked by the inclusion of melamine or urea formaldehyde resins in the composition. The primed PET film is stated to exhibit enhanced adhesion to a variety of coatings applied thereto, including reprographic coatings.

While s me of th se and other polyester film primer layers ar effective in enhancing the adhesive qualities of PET film, it is important for the film manufacturer that scrap film made during production must be recyclable through the film-forming process. Scrap film is normally comminuted, melted, extruded into the form of pellets, mixed with fresh virgin polyester, remelted and re-fed to the film-forming extruder. Typically high temperatures may be encountered during such processing of PET reclaim film. Many of the primer compositions discussed above are not stable at such temperatures and tend to impart an undesirable yellow or black discoloration to finished oriented PET film containing significant amounts of such primed reclaim film, particularly after repeated passes through the extruder. Such is the case with the vinylidene chloride-containing polymers used as PET primer layers and disclosed in U.S. Patent Nos. 2,627,088 and 2,698,240. It is also the case with primer layers based on copolymers containing vinyl chloroacetate as disclosed in U.S. Patent No. 3,674,538. It has been found that discoloration and degradation of these primer layers during the reclaim process is most likely attributable to the evolution of chlorine gas or hydrogen chloride in the case of chlorine-containing primer layers.

Another known primer is the thermoset, acrylic or methacrylic coatings taught in U.S. Patent No. 3,819,773, which can be applied to the PET film from aqueous medium. Such a primer layer enhances the adhesion of organic solvent based reprographic and drafting layers applied thereto. This patent also discloses that film primed with the acrylic coatings described therein may be reclaimed in the film forming extruder by mixing it with 50 percent by weight or more of virgin polyester and refeeding the mixture to the film-forming extruder. This reference discloses that problems of discoloration or degradation caused by certain prior art primer coatings are reduced. While this is true in comparison with the chlorine-containing primers discussed above, the acrylic primer coatings of U.S. Patent No. 3,819,773, which are crosslinked using the resinous crosslinking agents disclosed therein, still are found to give rise to an undesirable yellowing of the finished film containing

such primed film as reclaim, particularly when compared to film based solely on virgin polymer.

Summary of the Invention

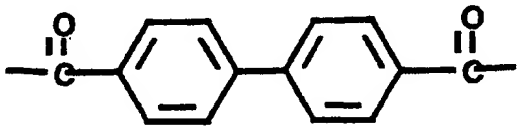
It has now been found that a primer coated biaxially oriented self-supporting mono- or multilayer copolyester film, wherein the copolyester is PENBB and wherein the primer coating contains an acrylic polymer provides excellent UV resistance, good dimensional stability, heat stability and hydrolysis resistance, lower moisture absorption, improved stiffness (tensile strength and modulus), while providing excellent adhesion to many organic solvent based coatings subsequently applied thereto, and reclaimability when reprocessed during the manufacture of the PENBB film while giving rise to hardly any degradation of the film.

Most of the acrylic coating compositions mentioned herein are disclosed in U.S. Patent No. 4,571,363.

U.S. Patent No. 3,008,934 discloses copolyesters containing as acid derived units 4,4'-bibenzoate and a host of other dicarboxylates including 2,6-naphthalic dicarboxylate. It also discloses oriented fibers and films prepared from these copolyesters, however, biaxially oriented PENBB films are not disclosed or envisioned. In particular, those films with improved stiffness (tensile modulus) and tensile strength in both MD and TD as well as thermostability, UV stability, hydrophobicity, dimensional stability and impermeability toward gases in comparison to PET film are not disclosed in U.S. Patent No. 3,008,934.

Description of the Preferred Embodiments of the Invention

PENBB as mentioned hereinbefore is a copolyester containing as acid-derived unit at least 5 mole percent of a radical of the formula



(bibenzoate, BB)

In the case that more than 10 mole percent of terephthalic acid derived radicals are present in the copolymer, the content of bibenzoate derived units is at least 25 mole percent. Films of these cop lyesters are mentioned in the unpublished German Patent Application P 4224161.8, which is incorporated herein by reference. Preferably PENBB is a copolyester wherein at least 80 mole percent of the acid derived units (NBB) consist of bibenzoate (20 to 80 mole percent, preferably 40 to 60 mole percent) and naphthalate (80 to 20 mole percent, preferably 60 to 40 mole percent). The remaining 20 or less mole percent may consist of other acid derived units, which e.g. affect the melting point or the crystallization kinetics. Preferably at least 80 mole percent of the diol-derived units consist of $-O(CH_2)_2-O-$units. The remaining 20 or less mole percent consist of other diol-derived units, which e.g. may also affect the melting point or the crystallization kinetics. It may also be desirable to replace minor amounts of the acid- and/or diol-derived units with hydroxycarboxylic-acid-derived units, e.g. such derived from p-hydroxybenzoic acid.

To produce the film, the polymer melt is extruded through a die onto a chill roll where it solidifies, is then biaxially oriented, heat set, optionally post treated and wound on a roll. In order to achieve the desired mechanical properties in the biaxially oriented PENBB film it is recommended that the IV value (inherent viscosity, as measured in a 1 : 1 weight-ratio mixture of pentafluorophenol and hexafluoroisopropanol at a concentration of 0.2 g/dl and a temperature of 25 °C) of the PENBB polymer after extrusion be > 0.5 dl/g and preferably > 0.55 dl/g. Biaxial drawing is performed such that the birefringeance is < 0.2, preferably < 0.1 to ensure adequately isotropic properties. Birefringeance as mentioned herein is the absolute value of the difference between the maximum and minimum refractive indices in the plane of the film, as measured on common instruments such as Abbé refractometer, optical bench or compensators. For a multilayer film known methods for coextrusion, in-line or off-line coating can be used. The solidified film as extruded on the chill roll should be obtained in an essentially amorphous state.

To achieve this, the melt film must be pinned to the chill roll by a known method such as electrostatic pinning or vacuum, air knif or the like.

The biaxial orientation of the film is achieved by stretching the film at elevated temperature in the machine (MD) and transverse direction (TD). This stretching can be either simultaneous or sequential. In the case of sequential stretching, the first stretching step can be in either MD or TD, followed by stretching in the other direction. The orientation in MD can also be achieved in several steps, either one after another prior to stretching in TD, or before and after the TD stretching. Preferred temperatures for stretching lie between the glass transition temperature and about 30°C above the cold crystallization temperature of the PENBB copolymer composition in use (both temperatures can easily be measured on amorphous films by DSC). The total stretch ratios (λ) in MD and TD lie between 1 : 2 and 1 : 10, preferably between 1 : 2.5 and 1 : 5. The product of the total stretch ratios should be between 1 to 30, and preferably between 5 to 20.

In order to optimize properties such as shrinkage, relaxation steps can be included in the orientation and heat setting processes.

The heat setting takes place at a temperature between the cold crystallization temperature and the melt temperature of the copolymer composition.

In some cases a surface treatment such as corona, plasma or flame treatment should be employed before winding the film on a roll.

Prior to coating the PENBB film surface with the acrylic coating composition, the film may be surface treated in a conventional manner by exposure to, e.g. an electric corona, plasma or flame treatment. Electric corona discharge is a conventional surface treatment which is commonly performed on a film to enhance its surface qualities, especially its adhesive and printing properties. Electric corona discharge methods and apparatus are described in U.S. Patent Nos. 3,057,792 and 4,239,973. If the surface treatment followed by the coating occurs before stretch orientation, heating the film before stretch orientation will drive off the water. For biaxially oriented film, prefera-

bly the corona treatment followed by the acrylic polymer coating may occur during the in-line manufacturing pr cess, either before stretch orientation, r between the machine draw and transverse draw of biaxial stretch orientation, or after stretch orientation. If the surface treatment and coating steps occur after stretch orientation, it is necessary to completely dry the film before winding. If the surface treatment and coating occur before orientation, or between draw during orientation, the later orientation steps which require the film to be heated would drive off the excess water from the acrylic polymer coating. Preferably, the corona treatment and subsequent acrylic polymer coating occur between draws during the stretch orientation step.

The mono- or multilayer biaxially oriented PENBB film is coated on the corona, plasma or flame treated surface preferably with an aqueous dispersion of the acrylic coating composition described below. The coating composition may conveniently be applied as an aqueous dispersion or emulsion using any of the well known coating techniques. For example, the film may be coated by roller coating, spray coating, gravure coating, or slot coating. The heat applied to the film during the subsequent pre-heating, stretching, and heat setting stages is generally sufficient to evaporate the water and crosslink the acrylic coating, if a crosslinkable monomer comprises a portion of the acrylic coating.

The coated, biaxially oriented PENBB film may then be heat treated for a period of time necessary to crystallize the film. Crystallization imparts the improved dimensional stability and excellent tensile properties to the PENBB film. The so coated, crystallized and biaxially oriented PENBB film is then wound onto a roll.

The above description describes an aqueous acrylic coating applied in-line (during manufacture). While this is the preferred process for the biaxially oriented copolyester film, organic solvent-based acrylic coatings as well as off-line coating (after manufacture of the film is complete) is a further aspect of the invention.

The acrylic copolymers used as primer layers according to this invention consist essentially of at least about 50 percent by weight of one or more

polymerized acrylic and/or methacrylic monomers. Although not necessary, the acrylic copolymer may contain from about 1 to 15 perc nt by weight of a copolymerizable comonomer, which in its copolymerized state is then capable of intermolecular crosslinking by the application of heat with or without the addition of a separate resinous crosslinking agent.

The acrylic component of the primer copolymers is preferably present at a level of from about 50 to 99 percent by weight and preferably comprises an ester of acrylic acid or methacrylic acid, especially an alkyl ester wherein the alkyl group contains up to 10 carbon atoms such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, t-butyl, hexyl, heptyl and n-octyl. Acrylic copolymers derived from a lower alkyl (C_1 to C_4) acrylate, especially ethyl acrylate, together with a lower alkyl methacrylate provide particularly effective adhesion between the PENBB film and superimposed reprographic or matte layers. Primer copolymers comprising an alkyl acrylate, e.g. ethyl acrylate or butyl acrylate, together with an alkyl methacrylate, e.g. methyl methacrylate, desirably in a total proportion in the range of about 70 to 95 weight percent, are particularly preferred. The acrylate comonomer of such acrylic/methacrylic combinations is preferably present in a proportion in the range of 15 to 65 mole percent and the methacrylate comonomer preferably in a proportion exceeding that of the acrylate comonomer generally by an amount in the range of 5 to 20 mole percent. The methacrylate is preferably present in a proportion in the range of 35 to 85 mole percent.

Examples of suitable crosslinking comonomers include N-methylol acrylamide, N-methylol methacrylamide and their corresponding ethers; epoxide materials such as glycidyl acrylate, glycidyl methacrylate and allyl-glycidyl ether; carboxyl containing monomers such as crotonic acid, itaconic acid or acrylic acid; anhydrides such as maleic anhydride or itaconic anhydride; hydroxy containing monomers such as allyl alcohol and hydroxy ethyl or propyl acrylate or methacrylate; amides such as acrylamide, methacrylamide or maleamide; and isocyanates such as vinylisocyanate or allylisocyanate.

Preferred comonomers from the above are N-methylolacrylamide and N-methylolmethacrylamide, mainly because copolymer chains containing one

of these monomers are capable of condensing with one another with the application of heat to form the desired inter-molecular crosslinking. In the case of copolymers containing the other functional monomers, it is necessary to form blends of two or more copolymers containing different functional comonomers to achieve the desired crosslinking, e.g. blending an acrylic/crotonic acid copolymer with an acrylic copolymer containing isocyanate, epoxide or N-methylol functionality capable of reacting with acidic functional groups.

Specific additional combinations of such blended acrylic copolymers include copolymers containing monomers having epoxide functionality combined with copolymers containing monomers having amine, acid anhydride, carboxyl, hydroxyl, or N-methylol functionality; copolymers containing monomers having N-methylol or N-methylol ether functionality combined with copolymers containing monomers having isocyanate functionality combined with copolymers containing monomers having carboxyl or hydroxyl functionality; and so forth. Preferably, the functional monomers present in the mixed copolymer systems are present in approximately equimolar amounts.

The acrylic copolymers also may contain up to about 49 percent by weight of one or more halogen-free non-acrylic monoethylenically unsaturated monomers interpolymerized therewith. Suitable comonomers include dialkyl maleates such as dioctyl maleate, di-isooctyl maleate and dibutyl maleate; vinyl esters of versatic acid; vinyl acetate, styrene, acrylonitrile and similar materials.

The preferred crosslinking mixed copolymer compositions for the purposes of this invention are approximately 50/50 blends of an ethyl acrylate/methyl methacrylate/crotonic acid copolymer with an ethyl acrylate/methyl methacrylate/glycidylacrylate copolymer, blends of an ethyl acrylate/methyl methacrylate/methacrylamide copolymer with an ethylacrylate/methyl methacrylate/N-methylolacrylamide copolymer; and compositions based on copolymers of ethyl acrylate/methyl methacrylate/N-methylolacrylamide such as copolymers containing from about 50 to 99 percent by weight of acrylic and/or methacrylic monomers, 0 to 49 percent by weight of the mono-ethylenically unsaturated monomer and about 1 to 15 percent by weight N-methy-

lolacrylamide. More preferred as copolymers containing from about 70 to 90 percent by weight of acrylic and/or methacrylic monomers, about 0 to 25 percent by weight of the mono-ethylenically unsaturated monomer, and from about 5 to 10 percent by weight N-methyolacrylamide.

As pointed out above, the polymeric acrylic primer composition does not require the presence of an external crosslinking agent such as a melamine or urea/formaldehyde condensate.

The primer coating is applied to the base mono- or multilayer PENBB film preferably as an aqueous dispersion or latex and at a solids concentration within the range of about 0.5 to 15 percent, preferably about 3 to 10 percent by weight. The preferred solids level is such as to yield a final dry coating thickness within the range of about 1 nm to 300 nm, which translates into a solids level on a dry weight basis of from 1 mg/m^2 to 300 mg/m^2. The preferred thickness range of the dried PENBB copolyester primer is from 15 to 50 nm, with 25 nm being the target thickness. The thickness of the base mono- or multilayer film to which the coating is applied may range from about 1 to about 250 μm.

The coating may be applied to one or both sides of the mono- or multilayer PENBB film, or it may be applied to one side only with optionally a different coating applied to the opposite side. Additives known in the art may be present in the coating formulation such as antistatic agents, wetting agents, surfactants, pH regulating agents, anti-oxidants, dyes, pigments, slip agents such as colloidal silica, and the like. Normally, it is desirable to include a surfactant to enhance the wettability of the aqueous coating to the base PENBB film.

PENBB film primed with the above acrylic composition has excellent utility as a film base for the production of photosensitive reprographic films. Such films are prepared by forming a coating on a surface of the primed PENBB film of a photosensitive composition comprising an organic solvent solution of a resinous binder containing or impregnated with a light-sensitive diazionium compound, and drying said coating. Resinous binders suitable for this purpose include cellulose acetate butyrate, cellulose acetate, cellulose

acetate propionate as well as vinyl polymers such as polyvinyl acetate. Suitable solvents include acetone, methyl ethyl ketone, methyl isobutyl ketone, ethylene glycol monomethyl ether and mixtures thereof. These reprographic coatings and their method of application and use are well known in the art.

Similarly the primed PENBB film of this invention forms an excellent substrate for the application of matte coatings which render the film suitable for use as a drawing or drafting material. These matte coatings may be based on an organic composition comprising a resinous binder and a finely divided particulate material which serves as a "toothing agent." The resinous binder may include the resinous materials referred to above as well as acrylic or methacrylic resins. The organic solvents may also include those listed above. Particulate materials include finely divided (less than 10 μm particle size) clays or silica. Other ingredients such as thickeners or dispersing agents may also be present in such matte formulations. Similar matte formulations are disclosed for example in British Patent No. 1,072,122 and U.S. Patent No. 3,624,021.

Yet another application of the primed film of this invention is as a packaging or label material. The primed biaxially oriented PENBB films demonstrate good adhesion to organic solvent based printing inks and markedly improved UV resistance, dimensional stability and stiffness (tensile strength) when compared with unprimed films, especially unprimed PET films. These inks may comprise organic solvent dispersions or solutions of pigments and/or dyes in combination with acrylic resins or other resins and thickening agents.

Accordingly, the excellent UV resistance, dimensional stability, stiffness (tensile strength) and adhesive qualities of PENBB film primed with the crosslinked acrylic copolymer layers of this invention to organic solvent based coatings applied thereto renders such film of more universal utility to the manufacturer of finished reprographic, graphic and packaging products.

Reprographic adhesion (Repro) is evaluated using a lacquer comprising (in parts by weight) 9 parts cellulose acetate butyrate of "20 second" grade

dissolved in a mixture of 88 parts ethylene glycol monomethyl ether (methyl cell s lve) and 3 parts methyl ethyl ketone with Rhodamine B dye added for color (3 parts of a 1 percent Rhodamine B dye in n-butanol). The lacquer is applied to the coated surface of the film by means of a wire-wound bar, Meyer rod No. 70, and cured in an oven for 5 minutes at 60°C. The coating is scored with a cross-hatched pattern by means of scalpel. A strip of adhesive tape (Scotch tape 610) is adhered to the cross-hatched area, rubbed with a fingernail to insure intimate contact and then pulled rapidly from the film. The amount of lacquer remaining in the cross-hatched area is expressed as a percentage of the amount remaining, i.e. no lacquer removed = 100 percent adhesion, all lacquer removed = 0 percent adhesion with intermediate adhesion values being assessed in terms of the proportion of lacquer area remaining adhered to the film. The test is done twice on two sheets (total of 4 areas tested) and the adhesion value reported as that of the test area showing the most failure.

Repro test results demonstrating less than 95 percent adhesion are not generally considered acceptable, whereas test results approaching or equaling 100 percent adhesion are the target standard.

The mechanical properties are measured in a tensile testing machine made by Zwick (Ulm, Germany) on 15 mm wide strips of film. The initial distance between the chucks is 100 mm and the crosshead speed is 100 mm/min for strength determination and 10 mm/min for the tensile modulus determination.

The UV resistance is tested by measuring the retention of tensile elongation after exposure to UV light in a "Suntest" apparatus manufactured by Heraeus (Hanau, Germany) for 14 days.

The following example is illustrative of the invention.

EXAMPLE

A latex comprising 4.5 percent by weight solids of a copolymer of 60 weight percent of methyl methacrylate, 35 weight percent ethyl acrylate, and 5 weight percent of N-methylolacrylamide, and a surfactant ar applied as a

primer coating to a PENBB film which is made of 289 parts by weight of dimethyl 2,6-naphthalene dicarboxylate, 322 parts by weight of dim thyl 4,4'-bibenzoate, 368 parts by weight of ethylene glycol. Granules of this polymer, having a melting point of 281°C are melted in a single screw extruder at temperatures of 280 to 320°C and extruded through a sheet die onto a cooling roll, temperature controlled at 30°C. A 120 μm thick film is obtained which is clear and transparent. Its density is 1.31 g/cm^3. This pre-film is then sequentially biaxially oriented. The longitudinally drawn film is corona treated by a corona discharge apparatus and thereinafter coated with the latex described above by reverse gravure coating. The corona treated longitudinally drawn, coated film is dried at a temperature of about 100°C. Thereafter the film is stretched in the transverse direction. The biaxially drawn film is heat set at a temperature of 230°C. These heat treatments result in the crosslinking of the primer coating.

The thus obtained primer coated PENBB film shows the following characteristics compared to a similarly coated PET film.

TABLE 1

Property	Units	PENBB	PET
Tensile Strength MD TD	(MPa)	240 180	100-200
Elongation At Break MD TD	(%)	25 20	50
Tensile Modulus MD TD	(GPa)	9.2 8.0	4-6
UV resistance	% retained tensile strength	77	0
Shrinkage (at 150°C for 15 minutes) MD TD	(%)	 0.3 0.3	 >1.0 >1.0

As is evident from the above table, stiffness (tensile strength, tensile modulus), elongation at break, shrinkage, and UV resistanc are improved as compared to similarly coated PET films. The film produced according to the above example is tested for reprographic adhesion as set forth above and provides greater than 95 percent adhesion as compared with a control uncoated polyester film which exhibited 0 percent adhesion.

THAT WHICH IS CLAIMED IS:

1. Primer coated, biaxially oriented self-supporting, mono- or multilayer copolyester film, wherein the copolyester is PENBB and wherein the primer coating contains an acrylic polymer.

2. Primer coated biaxially oriented copolyester film according to Claim 1, wherein the primer coating consists essentially of a copolymer of at least 50 percent by weight of polymerized acrylic and/or methacrylic monomers, from 1 to 15 percent by weight of a copolymerized functional group containing comonomer which in the copolymerized state is capable of inter-molecular crosslinking by the application of heat.

3. Primer coated biaxially oriented copolyester film according to Claim 1 or 2, wherein the primer coating contains from 0 to 49 percent by weight of one or more halogen-free mono-ethylenically unsaturated copolymerized monomers.

4. Claim 2 or 3, wherein said functional group present in said functional group containing monomer is selected from the group consisting of amine, amide, acid anhydride, N-methylol, carboxyl, hydroxyl, epoxy and isocyanate.

5. Primer coated biaxially oriented copolyester film according to Claim 2, 3 or 4, wherein said functional group containing comonomer is selected from the group consisting of N-methylolacrylamide and N-methylolmethacrylamide.

6. Primer coated biaxially oriented copolyester film according to any one or more of the preceding claims, wherein the coating is applied on one surface of the film.

7. Primer coated biaxially oriented copolyester film according to Claims 1-5, wherein the coating is applied on both surfaces of the film.

8. Primer coated biaxially oriented copolyester film a cording to any one or more f the prec ding claims, wh rein the film is a monolayer film.

9. Primer coated biaxially oriented copolyester film according to any one or more of the preceding claims with a birefringeance ≤ 0,2.

10. Primer coated biaxially oriented copolyester film according to any one or more of the preceding claims wherein the PENBB has an IV of ≥ 0.5 dl/g.

11. Use of the primer coated biaxially oriented copolyester film according to claim 1 as a packaging film.

12. Use of the primer coated biaxially oriented copolyester film according to claim 1 as a reprographic film.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/10699

A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) :B32B 27/06, 27/36
US CL :428/480, 482, 483, 353
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/480, 482, 483, 353

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 5,128,206 (FIARD ET AL) 07 JULY 1992. See entire disclosure.	1-3
Y	US, A, 4,556,606 (OLSON) 03 DECEMBER 1985. See entire disclosure.	1-3
Y	US, A, 4,571,363 (CULBERTSON ET AL.) 18 FEBRUARY 1986. See entire disclosure.	1-3
Y	US, A, 4,592,953 (FARRAR ET AL.) 03 JUNE 1986. See entire disclosure.	1-3

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be part of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E"	earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search	Date of mailing of the international search report
16 JANUARY 1993	26 FEB 1993
Name and mailing address of the ISA/ Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. NOT APPLICABLE	Authorized officer BLAINE COPENHEAVER Telephone No. (703) 308-2351

Form PCT/ISA/210 (second sheet)(July 1992)*

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/10699

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. [X] Claims Nos.: 11 & 12
because they relate to subject matter not required to be searched by this Authority, namely:

PCT Rule 39.1. Claims 11 and 12 are use- claims not written in method language as required.

2. [ ] Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. [X] Claims Nos.: 4 to 10
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. [ ] As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. [ ] As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. [ ] As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. [ ] No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention frist mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

[ ] The additional search fees were accompanied by the applicant's protest.

[ ] No protest accompanied the payment of additional search fees.